of the filler; ρ_f, density of the liquid; ρ_s, density of the filler; c_f, specific heat capacity of the liquid; c_s, specific heat capacity of the filler; p, pressure, p_c, capillary pressure; p_f, pressure of the liquid at the filler interface; p_0, air pressure in the pores of the filler; T_f, temperature at the filler interface; T_0, air temperature in the pores; μ_f, μ_0, viscosity of the liquid at the temperatures T_f, T_0, respectively; j_f, j_0, initial pressure gradients at these temperatures; $2l$, thickness of the impregnated layer; $\delta(x)$, thickness of the impregnated part of the working section of the apparatus.

LITERATURE CITED

1. Yu. A. Buevich and V. A. Kalinnikov, "Impregnation of a heated filler with a viscous liquid," Inzh.-Fiz. Zh., 36, No. 6, 997-1004 (1979).
2. A. Kh. Mirzadzhanzade, Problems of the Hydrodynamics of Viscoplastic and Viscous Liquids in Applications in Petroleum Extraction [in Russian], Aznefteizdat, Baku (1959).
3. L. Ya. Kosachevskii, E. A. Kosachevskaya, and V. M. Sidorova, "Selection of the optimum regime of the technological process of vacuum-forced impregnation," Inzh.-Fiz. Zh., 42, No. 5, 787-791 (1982).

MASS TRANSFER IN A REACTIVE SPHERICAL PARTICLE

Yu. A. Buevich and A. V. Kamennykh

UDC 541.128

A macrokinetic model is presented for the solid-phase transformation of a particle under arbitrary relationships between the sorption—desorption, dissolution, and diffusion rates of the initial and final products.

The problem of mass-transfer kinetics proceeding within a spherical particle during chemical or phase transformation occurs in connection with the necessity to model numerous technological processes of treating crushed and powdery materials. Theoretical information available in the literature on this subject refers primarily to situations when the macrokinetics of the process is constrained by the rate of just some one slowest stage, absorption external or internal diffusion, etc. [1, 2]. By using the model and method in [3], a theory is constructed in this paper in which it is assumed that the rates of several stages can be of comparable magnitude. Known simplified models turn out to be particular modifications of that proposed.

We make the following assumptions, analogous to those in [3]. The system has spherical symmetry. The mixing process in the external gas or liquid phase, as well as intrinsic transformation ("reaction"), are realized considerably more rapidly than the processes of adsorption and desorption from the external phase and the solid solution and diffusion in the solid phase. Upon compliance with these assumptions the reaction is localized in a very narrow spherical zone separating the domains in which the reaction has still not started or has already terminated, while the external phase characteristics at the particle surface are the same as far away from it.

Most important in principle is the assumption about the existence of a narrow reaction zone (a "front"). This assumption can be spoiled if the reaction rate is less than or on the order of magnitude of the rate of the mass-transfer processes. The reaction then proceeds simultaneously over the whole particle volume and the mathematical formulation of the problem is complicated substantially; at the present time only solutions obtained on the basis of approximate models, for which examples can be found in [4], are possible. Such a situation is especially characteristic for heterogeneous transformations in porous particles, although the representation of a reaction front in this case often turns out to be adequate (see, e.g., the problem of drying a porous particle in [5]).

For simplicity, we assume that there is just one substance ("reagent") that comes to the front from the external phase, and a single substance ("reaction product") that results in the

---

A. M. Gor'kii Ural State University, Sverdlovsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 48, No. 5, pp. 796-803, May, 1985. Original article submitted April 29, 1984.

0022-0841/85/4805-0580$09.50

long run. The kinetics equations for filling of the particle surface layer under the assumption of independent (Langmuir) adsorption are:

$$\frac{dS}{dt}=\alpha(1-S-S')-\beta S+\frac{\delta S}{\delta t},$$
$$\frac{dS'}{dt}=-\beta' S'+\frac{\delta S'}{\delta t}, \tag{1}$$

where $\delta S/\delta t$ and $\delta S'/\delta t$ are the rates at which the reagent and the reaction product fill this layer due to exchange with the solid solution.

The surface concentration of the reagent and the reaction product can be written as S/l^2 and S'/l'^2, respectively. The kinetics equations for exchange of the surface layer with the solid phase have the form [3]

$$\frac{\delta f}{\delta t}=-k_1 f+k_2 l(1-S-S')c_0=-D\left.\frac{\partial c}{\partial r}\right|_{r=R_0},$$
$$\frac{\delta f'}{\delta t}=-k_1' f'+k_2' l'(1-S-S')c_0'=-D'\left.\frac{\partial c'}{\partial r}\right|_{r=R_0}, \tag{2}$$
$$c_0=c|_{r=R_0},\quad c_0'=c'|_{r=R_0}.$$

The rates of reagent and reaction product dissolution in the solid phase are here considered proportional to the corresponding surface concentrations, while the sorption rates from the solid solution are proportional to the corresponding volume concentrations near the surface layer. In contrast to [3], it is here taken into account that only part of this layer which is not occupied by the reagent and the reaction product is accessible for filling.

Analogously to [3], on the reaction front we have boundary conditions and a material balance equation that govern the rate of front displacement

$$c=0,\quad -D\frac{\partial c}{\partial r}=\nu D'\frac{\partial c'}{\partial r},\quad r=R(t);$$
$$\varphi\frac{dR(t)}{dt}=D\left.\frac{\partial c}{\partial r}\right|_{r=R(t)}. \tag{3}$$

Neglecting the change in volume of the solid body, as a result of the reaction the quantity φ equals the product of the homogeneous reagent concentration in the solid-phase composition by the appropriate stoichiometric factor. Since usually $\varphi \gg c_0$, the relaxation time of the concentration fields of the diffusing reagent and reaction product is considerably less than the characteristic time of front advancement [3]. Consequently, it is admissible to utilize quasistationary representations for the mentioned fields in the range between $R = R(t)$ and R_0, which have the form

$$c=c_0\left(\frac{1}{R}-\frac{1}{R_0}\right)^{-1}\left(\frac{1}{R}-\frac{1}{r}\right),$$
$$c'=c_0'+\frac{D}{\nu D'}c_0\left(\frac{1}{R}-\frac{1}{R_0}\right)^{-1}\left(\frac{1}{r}-\frac{1}{R_0}\right),$$
$$c_0=k_1\left[k_2 l(1-S-S')+\frac{D}{R_0(R_0R^{-1}-1)}\right]^{-1}f, \tag{4}$$
$$c_0'=[k_2' l'(1-S-S')]^{-1}\left[k_1' f'+\frac{Dc_0}{\nu R_0(R_0R^{-1}-1)}\right].$$

The second equalities in (2) and the boundary conditions from (3) are used here. After manipulation we obtain the following system of equations from (1)-(4):

$$\frac{dS}{dt}=\alpha(1-S-S')-\beta S-\frac{k_1 D}{k_2 l R_0(R_0R^{-1}-1)(1-S-S')+D}S,$$
$$\frac{dS'}{dt}=-\beta' S'+\frac{k_1 D}{k_2 l R_0(R_0R^{-1}-1)(1-S-S')+D}\,\frac{l'^2}{\nu l^2}S,$$

$$\frac{dR}{dt}=-\frac{k_1D}{k_2lR_0(R_0R^{-1}-1)(1-S-S')+D}\left(\frac{R_0}{R}\right)^2\frac{S}{\varphi l^2}. \tag{5}$$

In the dimensionless variables and parameters

$$\xi=\frac{R}{R_0},\ \tau=\alpha t,\ \begin{Bmatrix}b\\ b'\end{Bmatrix}=\frac{1}{\alpha}\begin{Bmatrix}\beta\\ \beta'\end{Bmatrix},\ \varkappa=\frac{l'^2}{\nu l^2},$$
$$K=\frac{k_1}{\alpha},\ L=\frac{k_2lR_0}{D},\ M=\frac{1}{R_0\varphi l^2} \tag{6}$$

this system takes the form

$$\frac{dS}{d\tau}=1-(1+b)S-S'-\frac{KS}{L(\xi^{-1}-1)(1-S-S')+1},$$
$$\frac{dS'}{d\tau}=-b'S'+\frac{\varkappa KS}{L(\xi^{-1}-1)(1-S-S')+1}, \tag{7}$$
$$\frac{d\xi}{d\tau}=-\frac{KM}{L(\xi^{-1}-1)(1-S-S')+1}\frac{S}{\xi^2}.$$

For definiteness, we take

$$S=S'=0,\ \xi=1,\ \tau=\tau_0\geqslant 0, \tag{8}$$

as initial conditions, where the quantity τ_0 characterizes the duration of the induction period (the build-up of a spherically symmetric reaction front).

In the general case the solution of the system (7) under the conditions (8) or any other initial conditions is not expressed in analytic form although it can easily be obtained numerically. But for a number of important particular cases simplified analytical relationships, adequate for the description of many real processes, can be obtained from (7) and (8). The time-dependence of the degree of transformation $A = 1 - \xi^3$ and its rate of change dA/dt is of special interest.

For small τ we obtain approximately

$$S\approx\tau-\tau_0,\ S'\approx(\varkappa K/2)(\tau-\tau_0)^2,\ \xi\approx[1-(3KM/2)(\tau-\tau_0)^2]^{1/3},$$
$$A\approx\frac{3\alpha k_1(t-t_0)^2}{2R_0\varphi l^2},\ \frac{dA}{dt}\approx\frac{3\alpha k_1(t-t_0)}{R_0\varphi l^2} \tag{9}$$

(the quantities A and dA/dt are expressed in the original dimensional variables). Therefore, in the initial stage of the process the transformation rate is inversely proprotional to the linear size of the particle and is constrained only by adsorption and by dissolution of the reagent in the solid materials, but is independent of the rates of all other mass-transfer processes under consideration, as should have been expected.

Let us consider the quasistationary mode corresponding to a slow change in the fractions of surface layer area filled by the reagent and the reaction product. Mathematically this means that the derivatives in the left sides of the first two equations in (7) are small compared with the terms in the right sides of these equations. The quasistationary mode can be realized in different bands of values of the dimensionless parameters (6). We briefly investigate the simplest possible variations.

For instance, let $K \ll 1 + b$, $b' \gtrsim 1$ (the rate of reagent dissolution is small compared with the rates of reagent exchange between the surface layer and the external phase, the rate of desorption of the reaction product in the external phase is of the same order of magnitude or greater than the rate of reagent adsorption therefrom). Then in a lapse of dimensionless time exceeding the quantity $(1 + b)^{-1}$ several times, a quasistationary mode is built up after the beginning of the process in which $S \approx (1 + b)^{-1}$, $S' \approx 0$. Integrating the last equation in (7) under the initial condition on ξ from (8), we obtain the following relationship after simple manipulations

$$1-\frac{2}{3}\left(1-\frac{1+b}{bL}\right)A-(1-A)^{2/3}\approx\frac{2KM}{bL}\tau\quad(\tau\gg\tau_0). \tag{10}$$

This relationship determines the dependence of the degree of transformation on the time and the parameters and generalizes the known Ginstling–Brownstein formula [6] that is obtained from (10) if the coefficient for A equals 2/3. For this $bL \gg 1 + b$ is evidently necessary; this strong inequality is not satisfied in important cases when $k_2 l \lesssim D/R_0$ (the effective rate of reagent sorption from the solid phase is of the same order of magnitude or less than the rate of its diffusion in it) or for $L > 1$; $\beta \lesssim \alpha$ (the rate of reagent desorption in the external phase is less than or on the order of the rate of adsorption). For spoilage of the mentioned inequality the Ginstling–Brownstein formula is inapplicable to the description of the transformation kinetics; in the limit case $bL \ll 1 + b$ we have from (10)

$$A \approx \frac{3KM}{1+b}\tau. \tag{11}$$

Now let $\xi \gg L(1 + L)^{-1}$, as is possible in a broad range of values of ξ or A for $L \ll 1$. In this case a quasistationary mode is built up in which $S \approx [1+b+(1+\varkappa/b')K]^{-1}$, $S' \approx (\varkappa/b')KS$, and we obtain in place of (10) after calculation

$$A \approx \frac{3KM}{1+b+(1+\varkappa/b')K}\tau. \tag{12}$$

This formula generally has nothing in common with the Ginstling–Brownstein formula; for $K \ll 1 + b$, $b' \gtrsim 1$ it goes over into (11). Formulas (11) and (12) correspond to a constant transformation rate; in dimensional variables we obtain from (12)

$$\frac{dA}{dt} \approx \frac{3}{R_0\varphi}\,\frac{\alpha k_1}{l^2(\alpha+\beta+k_1)+l'^2(\alpha/\nu\beta')k_1}. \tag{13}$$

The influence of the separate elementary mass-transfer process rates on the transformation rate is hence clarified especially clearly. Thus for a relative increase in the rate of reagent adsorption, dA/dt tends to the characteristic limit value $3k_1/R_0\varphi l^2$, i.e., is determined just by the rate of reagent dissolution in the solid material, for a relative increase in the rate of dissolution, dA/dt tends to $(3\alpha/R_0\varphi l^2)(1+l'^2\alpha/\nu l^2\beta')^{-1}$, i.e., is determined by the rates of reagent adsorption from the external phase and desorption of the reaction product therein, etc. Separately, we note that the inhibiting influence of surface layer contamination by the reaction product on the transformation process is substantial if the rate of desorption of this substance in the external phase is quite small ($\beta' \ll \alpha$). In this case we have the limit formula $dA/dt \approx 3\beta'/\nu R_0\varphi l'^2$.

We obtain still another relationship for the concluding stage of the process when $\xi \ll bL/K$. In this case $S \approx (1 + b)^{-1}$, $S' \approx 0$, and in dimensional variables

$$A \approx 1 - \left[1 - \frac{2\alpha k_1 D}{R_0^2\varphi l^3\beta k_2}\,(t-t^*)\right]^{3/2}, \tag{14}$$

where t^* is a certain characteristic time of the beginning of the concluding stage which plays the part of a constant of integration in (14). Therefore, the rate of transformation in the concluding stage is approximately determined in equal degree by the rates of all the reagent sorption–desorption processes as well as its diffusion, and decreases monotonically to zero at a certain time $t = t_*$.

If it is assumed that the duration of the whole transformation process is determined mainly by the length of its concluding stage ($t_* \gg t^*$), as is quite probable for $bL \gg K$, then we obtain and estimate for the time t_* of completion of the mentioned process from (14):

$$t_* \approx \frac{R_0^2\varphi l^3\beta k_2}{2\alpha k_1 D}. \tag{15}$$

Exact values of t_* for different values of the kinetic and modal parameters with the initial substantially nonstationary stages of the process taken into account can be obtained by numerical integration of (7). Characteristic dependences of this time on the parameters are shown in Fig. 1.

Therefore, the approximate Ginstling–Brownstein, Yander, and other formulas known from the literature [1, 2] are easily obtained from the proposed theory in particular cases. However, the domains of adequacy of these formulas are quite specific, and in many important cases are insufficient to describe real heterogeneous processes, investigation of appropriate

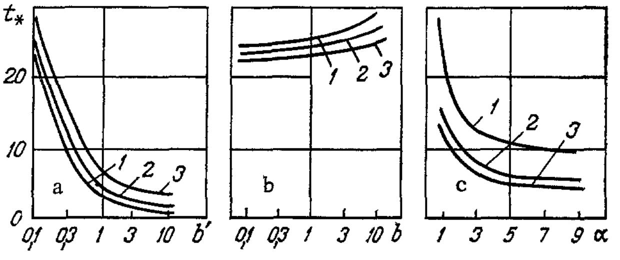

Fig. 1

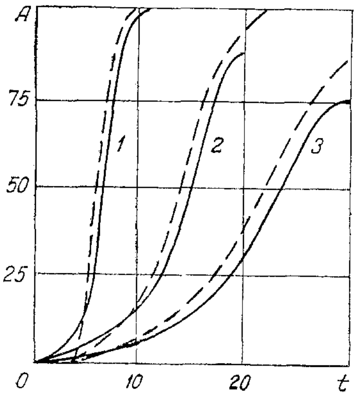

Fig. 2

Fig. 1. Dependence of the time of transforamtion t_*, min, on the parameters: a) $\alpha = 1$ min^{-1}, $K = 5$, $L = 1$, $M = 0.3$, $\varkappa = 3$, 1-3) $c = 0.1$; 1; 10; b) $L = 1$, $M = 0.3$; $\varkappa = 3$; 1-3) $\alpha = 1$, 3, 5 min^{-1}, $K = 10$; 3.33; 2, $b' = 0.1$; 0.033; 0.02; c) $L = 1$; $M = 0.1$; $\varkappa = 3.3$; 1-3) for $\alpha = 10$ min^{-1} and $K = 1$; 0.5; 0.1.

Fig. 2. Kinetics of reduction of germanium dioxide by hydrogen for T = 600°C and pressures 709, 608, and 532 mm Hg (1-3); solid curves are experiment [7], dashed lines are theory ($L = 0.5$, $M = 1$, $\varkappa = 1$ for 709 mm Hg, $\alpha = 0.05$ min^{-1}, $K = 0.1$, $b = b' = 1$). A, %; t, min.

solutions of the problem (7), (8) is required. Analysis of numerical solutions of this problem indicates that the proposed theory permits giving a natural explanation to practically all the features of the experimental kinetic curves for different kinds of heterogeneous processes. Unfortunately, a detailed comparison of theory with the test data in the literature is made difficult by the fact that some of the kinetic coefficients in the theory are usually generally unknown for specific processes or are not contributed by the authors in their publications. Consequently, the theoretical and experimental curves were superimposed by a somewhat arbitrary selection of the values of the dimensionless parameters introduced with the accessible information about these processes taken into account.

For many real heterogeneous processes the time-dependence of the degree of transformation has the characteristic S-shape [1, 2]. Such curves are obtained from theory in a very broad range of parameter variation. For example, displayed in Fig. 2 are experimental curves obtained for the reduction of powdery germanium dioxide by hydrogen under different pressures [7], and the corresponding theoretical dependences. The values of the parameters dependent on the quantity α were calculated under the assumption of compliance with the Syverts law, i.e., it was considered that α is proportional to the square root of the hydrogen partial pressure. Curves of the type shown in Fig. 2 are characteristic for a situation with moderate rates of reagent adsorption and dissolution in particular (as a considerable induction period specifies) and high rates of reaction-product desorption in the external phase (which assures sufficient "purity" of the particle surface).

If the reagent adsorption and dissolution rates are relatively large, the rate of transformation reaches a maximum value very rapidly (such that it turns out to be impossible sometimes even to measure it in the initial stage of its growth). At this time the fraction of the surface layer area filled with reagent is close to one. The appearance of excess quantities of reaction product results in its partial filling of the mentioned layer, which together with the advancement of the front into the particle bulk contributes to a rapid drop in the transformation rate. The fraction of surface layer area filled by the reaction product grows if its rate of desorption is not too small. The drop in the transformation rate specifies a diminution in the diffusion flux of the reaction product to the surface, and then also a diminution in its filling of the surface layer. Under certain conditions this causes not only an increase in the filling of this layer by the reagent but also a new rise in the rate of transformation with the build-up of a mode with oscillations, where the possibility of its existence was mentioned in [3]. Experimental dependences of dA/dt on A are presented in Fig. 3 for the reduction of uranous oxide by methane [8], as are also curves following from the theory under consideration. It is seen that the appearance of maximums and minimums on the kinetic curves can easily be explained even without involving the additional reasoning,

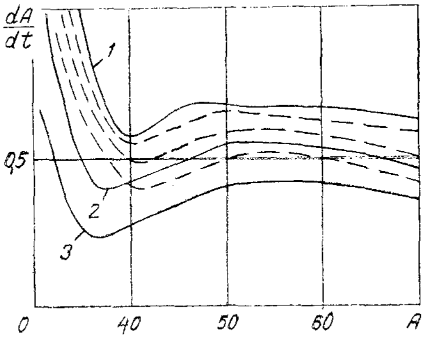

Fig. 3

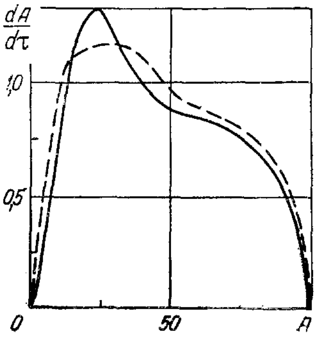

Fig. 4

Fig. 3. Kinetic curves of uranous oxide, uranium oxide reduction by methane for T = 600°C and the pressures 100, 200, and 300 mm Hg (1-3); solid curves are experiment [8], dashed curves are theory (L = 1; M = 0.3; $\varkappa$ = 3.3; 1-3) α = 7, 5, 4 min^{-1}, K = 0.71; 1.0; 1.25; b = b' = 0.43; 0.60; 0.75). dA/dt, min^{-1}.

Fig. 4. Characteristic dependence of the rate of nickel oxide reduction on the degree of reduction [9] and the theoretical curve (dashed) for K = 0.1, L = 1.0, M = 0.3, $\varkappa$ = 1.0, b = 1, b' = 0.2.

ordinarily utilized in interpreting test data, about the growth in oxide crystal lattice defects during transformation, etc.

Certain other observable features of the kinetic curves, for instance, the appearance of inflection points on the dependences of dA/dt on A, can also be explained by the effect of surface layer etching by the reaction product. One such dependence obtained in tests [9] is presented in Fig. 4 together with the appropriate theoretical curve.

The disadvantage of the widely known approximate Ginstling–Brownstein, Yander, and other formulas for an adequate description of many heterogeneous processes stimulated numerous attempts at generalization of these formulas, not all of which are error-free by far. As an example, we mention the recent paper [10] in which the advancement of a spherical reaction front in the simplest case is studied, when only reagent diffusion is essential (the Ginstling–Brownstein theory should be valid even in the quasistationary approximation).

After a number of erroneous reasoning and citations completely without proof for different situations of no relationship to that under examination, the authors of [10] arrive at the system of equations

$$\frac{dV}{dt}\sim\frac{dc}{d(R_0-R(t))},\quad \frac{dV}{dt}\sim V,\quad \frac{dc}{dt}\sim\text{const},\quad V=\frac{4\pi}{3}(R_0^3-R^3(t)),$$

where c(t) is some generally unknown function of just the time, which replaces (in calculating the gradient!) the true field of reagent concentration. Without going into the details of the argument in [10], we note that the system of these equations, even if the latter were true singly, is inwardly contradictory. The quantity R(t) is obtained in [10] by integrating the equation $dR(t) \sim -V^{-1}$ that follows from this system after c(t) is eliminated. A perfectly different result is obtained for R(t) if the second equation is integrated directly.

In conclusion, let us note that the results presented above are extended easily even to more complex situations when there are several reagents coming in from the external phases, several reaction products, and possibly several reaction going on in parallel. The boundary conditions on the front are again written in conformity with the stoichiometric equations for these reactions, in the fast-reaction approximation. Such an analysis of these processes is not the problem in this paper. Consequently, we here emphasize just that a qualitative explanation can hence be given successfully for many important phenomena and features of the heterogeneous processes observed in practice. For instance, the addition of a second gas-reducer in the external phase of a widespread process for reduction of powdery metal oxides (with the partial pressure of the first retained) cannot only not increase but also substantially diminishes the rate of the reduction process because of the selective sorption of the appropriate reaction product by the surface layer.

NOTATION

A, degree of transformation; b — β/α; c, concentration; D, diffusion coefficient; f, surface concentration; k_1, k_2, adsorption and desorption rate constants in the solid phase; l, linear dimension of the area occupied by one molecule; K, L, M, dimensionless parameters defined in (6); R, R_0, reaction front and particle radii, respectively; r, radial coordinate; S, fraction of surface layer filling; t, time; t_*, transformation time; α, adsorption rate constant from the gas phase; β, desorption rate constant in the gas phase; $\varkappa$ — $l'^2/\nu l^2$; ν, stoichiometric coefficient; ξ — R/R_0; τ — αt; τ_0, dimensionless duration of the induction period; φ, reagent concentration in the initial solid phase; the prime denotes quantities referring to the gaseous reaction product.

LITERATURE CITED

1. B. Delmon, Kinetics of Heterogeneous Processes [Russian translation], Mir, Moscow (1972).
2. P. Barre, Kinetics of Heterogeneous Processes [Russian translation], Mir, Moscow (1976).
3. Yu. A. Buevich, "Mass transfer in chemical or phase transformations in a solid," Inzh.-Fiz. Zh., 46, No. 2, 233-240 (1984).
4. V. A. Kirakosyan and A. P. Baskakov, "Mathematical model of heterogeneous reactions," Inzh.-Fiz. Zh., 43, No. 4, 589-597 (1982).
5. V. E. Babenko, Yu. A. Buevich, and N. M. Shepshuk, "Quasistationary mode of drying a spherical particle," Teor. Osn. Khim. Tekh., 9, No. 2, 274-277 (1975).
6. Yu. D. Tret'yakov, Solid Phase Reactions [in Russian], Khimiya, Moscow (1978).
7. G. A. Agarkova and R. M. Sadykov, "Kinetics of low-temperature reduction of germanium dioxide by hydrogen," Zh. Prikl. Khim., 44, No. 7, 1629-1632 (1971).
8. Yu. N. Semavin and V. G. Vlasov, "Interaction of uranous-uranium oxide with methane," Zh. Prikl. Khim., 42, No. 9, 1010-1014 (1969).
9. V. V. Veselov and V. P. Dorokhovich, "Low-temperature reduction of iron and other metal oxides by methane," Zh. Prikl. Khim., 41, No. 7, 1489-1493 (1968).
10. I. G. Kovzun and F. D. Ovcharenko, "On the kinetics of chemical interaction of gases and fluids with porous particles," Kolloidn. Zh., 44, No. 3, 465-470 (1982).

HEAT EXCHANGE BETWEEN A LAMINAR AND PULSATING PLASMA JET AND A BARRIER

J. Dundr, L. Kreichi,
V. L. Sergeev, A. S. Strogii,
and F. B. Yurevich*

UDC 533.951.7:536.24.01

The influence of the parameters of the plasma stream and their time variation on the intensity of heat exchange near the stagnation point of a blunt body is analyzed.

Technological processes based on the use of a low-temperature plasma are presently used in various branches of science and engineering. High-temperature gas streams are obtained by electric-arc heating of gas in plasmatrons. Since the main technological zone in the majority of plasma processes is the plasma jet generated by the heater, for its efficient practical use it is important to know the amount of the heat flux from the jet to the solid, which depends on the operating parameters of the plasmatron and the distance from the nozzle exit cross section to the barrier.

---

*Deceased.

Institute of Thermomechanics, Czechoslovakian Academy of Sciences, Prague. A. V. Lykov Institute of Heat and Mass Transfer, Academy of Sciences of the Belorussian SSR, Minsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 48, No. 5, pp. 803-809, May, 1985. Original article submitted April 11, 1984.

0022-0841/85/4805-0586$09.50